DTU Library

Extreme nonlinearities in InAs/InP nanowire gain media: the two-photon induced laser

Capua, Amir; Kami, Ouri; Eisenstein, Gadi; Reithmaier, Johanii Peter; Yvind, Kresten

Published in:
Optics Express

Link to article, DOI:
10.1364/OE.20.005987

Publication date:
2012

Document Version
Publisher's PDF, also known as Version of record

Extreme nonlinearities in InAs/InP nanowire gain media: the two-photon induced laser

Amir Capua,[1,*] Ouri Karni,[1] Gadi Eisenstein,[1] Johann Peter Reithmaier,[2] and Kresten Yvind[3]

[1]*Department of Electrical Engineering, Technion,Haifa,32000, Israel*
[2]*Institute of Nanostructure Technologies and Analytics,University of Kassel, Kassel, D-34132, Germany*
[3]*Department of Photonics Engineering, DTU-Fotonik, Technical University of Denmark, Lyngby, Denmark*
[*]*acapua@tx.technion.ac.il*

Abstract: We demonstrate a novel laser oscillation scheme in an InAs / InP wire-like quantum dash gain medium. A short optical pulse excites carriers by two photon absorption which relax to the energy levels providing gain thereby enabling laser oscillations. The nonlinear dynamic interaction is analyzed and quantified using multi-color pump-probe measurements and shows a highly efficient nonlinear two photon excitation process which is larger by more than an order of magnitude compared to common quantum well and bulk gain media. The dynamic response of the nonlinearly induced laser line is characterized by spectrally resolved temporal response measurements, while changes incurring upon propagation in the stimulating short pulse itself are characterized by frequency resolved optical gating (FROG).

OCIS codes: (140.5960) Semiconductor lasers; (250.5590) Quantum-well, -wire and -dot devices; (250.5980) Semiconductor optical amplifiers; (320.0320) Ultrafast optics; (320.2250) Femtosecond phenomena; (250.4390) Nonlinear optics, integrated optics.

References and links

1. P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, "Generation of optical harmonics," Phys. Rev. Lett. **7**(4), 118–119 (1961).
2. A. Hayat, P. Ginzburg, and M. Orenstein, "Observation of two-photon emission from semiconductors," Nat. Photonics **2**(4), 238–241 (2008).
3. J. Costello, "Extreme-ultraviolet sources: higher harmonics with plasmonics," Nat. Photonics **5**(11), 646–647 (2011).
4. F. Wang, R. Deng, J. Wang, Q. Wang, Y. Han, H. Zhu, X. Chen, and X. Liu, "Tuning upconversion through energy migration in core-shell nanoparticles," Nat. Mater. **10**(12), 968–973 (2011).
5. W. Wenseleers, F. Stellacci, T. Meyer-Friedrichsen, T. Mangel, C. A. Bauer, S. J. K. Pond, S. R. Marder, and J. W. Perry, "Five orders-of-magnitude enhancement of two-photon absorption for dyes on silver nanoparticle fractal clusters," J. Phys. Chem. B **106**(27), 6853–6863 (2002).
6. A. Capua, G. Eisenstein, and J. P. Reithmaier, "A nearly instantaneous gain response in quantum dash based optical amplifiers," Appl. Phys. Lett. **97**(13), 131108 (2010).
7. C. F. Zhang, Z. W. Dong, G. J. You, R. Y. Zhu, S. X. Qian, H. Deng, H. Cheng, and J. C. Wang, "Femtosecond pulse excited two-photon photoluminescence and second harmonic generation in ZnO nanowires," Appl. Phys. Lett. **89**(4), 042117 (2006).
8. C. Zhang, F. Zhang, T. Xia, N. Kumar, J. I. Hahm, J. Liu, Z. L. Wang, and J. Xu, "Low-threshold two-photon pumped ZnO nanowire lasers," Opt. Express **17**(10), 7893–7900 (2009).
9. S. Dayal and C. Burda, "Semiconductor quantum dots as two-photon sensitizers," J. Am. Chem. Soc. **130**(10), 2890–2891 (2008).
10. S. Schneider, P. Borri, W. Langbein, U. Woggon, J. Förstner, A. Knorr, R. L. Sellin, D. Ouyang, and D. Bimberg, "Self-induced transparency in InGaAs quantum dot waveguides," Appl. Phys. Lett. **83**(18), 3668–3670 (2003).
11. P. Aivaliotis, E. A. Zibik, L. R. Wilson, J. W. Cockburn, M. Hopkinson, and N. Q. Vinh, "Two photon absorption in quantum dot-in-a-well infrared photodetectors," Appl. Phys. Lett. **92**(2), 023501 (2008).
12. A. Capua, S. O'Duill, V. Mikhelashvili, G. Eisenstein, J. P. Reithmaier, A. Somers, and A. Forchel, "Cross talk free multi channel processing of 10 Gbit/s data via four wave mixing in a 1550 nm InAs/InP quantum dash amplifier," Opt. Express **16**(23), 19072–19077 (2008).
13. P. Day, K. Nguyen, and R. Pachter, "Calculation of one- and two-Photon absorption spectra of thiolated gold nanoclusters using time-dependent density functional theory," J. Chem. Theory Comput. **6**(9), 2809–2821 (2010).

#161161 - $15.00 USD Received 10 Jan 2012; revised 9 Feb 2012; accepted 9 Feb 2012; published 27 Feb 2012

14. S. A. Khan, D. Senapati, T. Senapati, P. Bonifassi, Z. Fan, A. K. Singh, A. Neeley, G. Hill, and P. C. Ray, "Size dependent nonlinear optical properties of silver quantum clusters," Chem. Phys. Lett. **512**(1-3), 92–95 (2011).
15. H. Dery, E. Benisty, A. Epstein, R. Alizon, V. Mikhelashvili, G. Eisenstein, R. Schwertberger, D. Gold, J. P. Reithmaier, and A. Forchel, "On the nature of quantum dash structures," J. Appl. Phys. **95**(11), 6103–6111 (2004).
16. A. Capua, G. Eisenstein, and J. P. Reithmaier, "Ultrafast cross saturation dynamics in inhomogeneously broadened InAs/InP quantum dash optical amplifiers," Appl. Phys. Lett. **98**(10), 101108 (2011).
17. H. Ju, A. V. Uskov, R. Nötzel, Z. Li, J. Molina Vázquez, D. Lenstra, G. D. Khoe, and H. J. S. Dorren, "Effects of two-photon absorption on carrier dynamics in quantum-dot optical amplifiers," Appl. Phys. B **82**(4), 615–620 (2006).
18. H. Dery and G. Eisenstein, "Self-consistent rate equations of self assembly quantum wire lasers," IEEE J. Quantum Electron. **40**(10), 1398–1409 (2004).
19. M. Kessler and E. P. Ippen, "Subpicosecond spectra gain dynamics in AlGaAs laser diodes," Electron. Lett. **24**(17), 1102–1104 (1988).
20. R. Trebino, *Frequency-Resolved Optical Gating: The Measurement Of Ultrashort Laser Pulses* (Kluwer Academic Publishers, 2002), Chap. 16.
21. A. Capua, A. Saal, O. Karni, G. Eisenstein, J. P. Reithmaier, and K. Yvind, "Complex characterization of short-pulse propagation through InAs/InP quantum-dash optical amplifiers: from the quasi-linear to the two-photon-dominated regime," Opt. Express **20**(1), 347–353 (2012).
22. D. Hadass, V. Mikhelashvili, G. Eisenstein, A. Somers, S. Deubert, W. Kaiser, J. P. Reithmaier, A. Forchel, D. Finzi, and Y. Maimon, "Time-resolved chirp in an InAs/InP quantum-dash optical amplifier operating with 10Gbit/s data," Appl. Phys. Lett. **87**(2), 021104 (2005).

1. Introduction

The quest for materials with ever larger optical nonlinearities has been in the forefront of research since discovery of optical harmonics [1]. The highly non centro-symmetric structure of semiconductor quantum dots and quantum wires enables potentially large nonlinear responses while offering integration in applications with demanding requirements on the nonlinear optical interaction.

An increasing number of experimental observations have demonstrated the large potential of nonlinear nanostructure-based devices [2–5] and of nano-scale semiconductors in particular [6–12]. Embedding such materials, which exhibit highly localized fields and large two-photon absorption (TPA) cross sections [13–14], in a waveguide can further enhance their nonlinear properties as the interaction length is extended.

An important class of semiconductor nano-materials is quantum dashes (QDash) which enable optical gain in the important wavelength range of 1550 nm. QDashes are a dense assembly of quantum wires [15], which have different sizes and hence exhibit an inhomogeneously broadened gain. Their basic microscopic structure leads to unique nonlinear properties where two-photon interaction excites carriers into the hosting quantum well (QWell) as well as directly into the high energy tail of the wire-like nanostructures [6].

Here we demonstrate an extremely efficient two-photon induced gain mechanism employed to initiate laser oscillations in a wire-like quantum dash gain medium. We show the two-photon interaction coefficient to be more than one order of magnitude larger than in bulk and quantum well gain media. Combined with multi-wavelength spectroscopy and temporal analysis of the laser line, a deep understanding of the nonlinear processes is obtained. Our work opens a new discipline for exotic nonlinear material research and design, and has significant implications to the realization of new classes of devices such as a true two-photon laser [2].

2. Experimental results

The experiments we describe were performed using an anti-reflection coated, 1.5 mm long, InAs QDash optical amplifier whose structure is described in [16]. The amplified spontaneous emission spectra are shown in Fig. 1(a). At low and moderate bias levels, the emitted spectra are broad as expected in an inhomogeneously broadened gain medium. However, at a high bias of 230 mA (which is 5.5 times the oscillating threshold prior to the anti-reflection facet coating), laser oscillations occur due to the finite residual facet reflectivity estimated to be 0.03% - 0.05%.

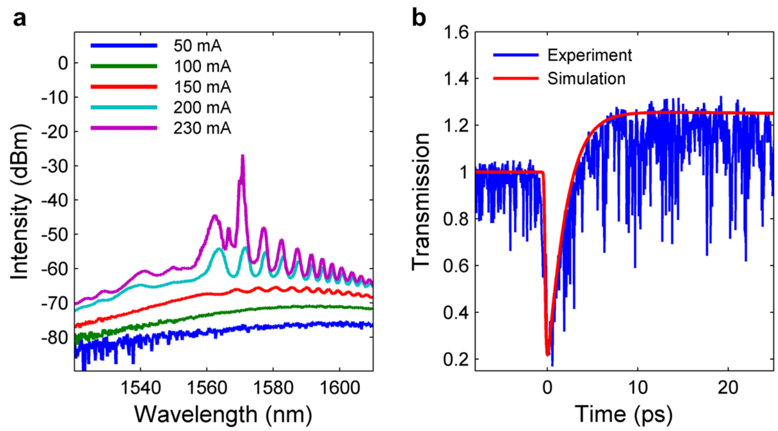

Fig. 1. Spectroscopic data. (a) Bias dependent amplified spontaneous emission spectra. (b) Multi-wavelength pump-probe response at a bias of 150 mA; pump at 1550 nm and probe at 1600 nm.

Dynamical nonlinear responses were characterized at moderate bias levels using a multi-wavelength pump-probe measurement set up whose details are described in [16]. Figure 1(b) describes the response of a CW probe tuned at 1600 nm to the perturbation of a 150 fs wide pump pulse centered at 1550 nm for a bias level of 150 mA. The pump pulse causes cross gain modulation and hence the gain at 1600 nm reduces. Most important however, the gain recovers after a few ps to a level higher than that prior to the arrival of the pump pulse. The generation of the extra carriers enabling the gain originates from an indirect process of two-photon excitation into a continuum of QWell states (whose band edge is located 150 meV above the QDash ground state) followed by relaxation and capture to the ground states of all QDashes as demonstrated by multi-wavelength pump-probe measurements in [6]. The highly populated QWells serve as a carrier reservoir and since their lifetime is long, the high gain at the ground state lasts for hundreds of ps. This is the first experimental observation of this kind which confirms precisely the theoretical prediction described in [17].

Quantification of the nonlinear TPA coefficient and the relevant dynamical time constants was obtained by solving a set of distributed dynamical equations coupling the electromagnetic (EM) field with an effectively three level carrier density similar to the ones used in [17].

A simplified version of the propagation equation, describes the photon evolution along the waveguide:

$$\frac{dS}{dz} = \Gamma_L g S - \Gamma_{NL} \beta S^2 \tag{1}$$

$$\beta \propto \left| \sum_k \frac{\mu_{ik}\hat{e}_1 \mu_{kf}\hat{e}_2}{E_k - E_1 + i\Gamma_k} + \sum_k \frac{\mu_{ik}\hat{e}_2 \mu_{kf}\hat{e}_1}{E_k - E_2 + i\Gamma_k} \right|^2 . \tag{2}$$

In Eq. (1), S is the photon density, Γ_L and Γ_{NL} are the transverse modal linear and nonlinear confinement factors, g is an effective gain coefficient which considers the inhomogeneous broadening and β is the nonlinear TPA coefficient. Equation (2) describes qualitatively the parameters which determine β where the summation occurs over the intermediate virtual states, k, having energies E_k. E_1 and E_2 are the energies of the two-photons while μ, $\hat{e}$, and Γ_k are the dipole moment of the initial to virtual and virtual to final states transitions, the field polarization and the decay rate, respectively.

For the specific QDash structure at hand, fitting the model to the experiments (Fig. 1(b)) yields an extracted QWell to QDash capture time of 1.3 ps and a 1 ns equivalent QWell carrier lifetime. Using confinement factors of 0.025 for both Γ_{NL} and Γ_L (calculated from geometrical considerations), a fit of Eq. (1) resulted in an extremely large TPA coefficient of

#161161 - $15.00 USD Received 10 Jan 2012; revised 9 Feb 2012; accepted 9 Feb 2012; published 27 Feb 2012

540 cm/GW, which is more than an order of magnitude larger than in conventional bulk and QWell semiconductor optical amplifiers testifying to the potential of the QDash gain media for nonlinear applications. The large coefficient stems mainly from a large surface area with a significant index contrast typical of the dense assembly of InAs dashes (wires) which are embedded within AlGaInAs. That index contrast induces large potential distortions which enhance, in turn, the dipole moment μ in Eq. (2). Moreover, in addition to the large TPA coefficient, the small nonlinear confinement factor extends the effective nonlinear interaction length thereby enhancing the two-photon excitation into the barrier.

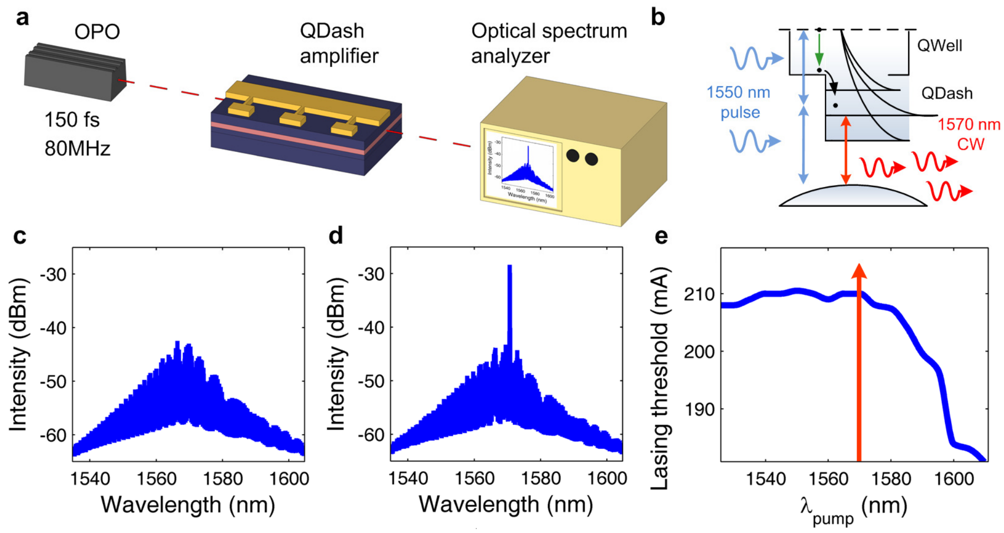

Fig. 2. Optically triggered laser oscillations. (a) Experimental schematic. (b) Conceptual description of band diagram together with the sequence of excitation and emission processes. (c) Output spectrum with no optical injection. (d) Output spectrum with optical injection. (e) Laser oscillation threshold dependence on optical pump wavelength. The oscillating wavelength, 1570 nm, is marked artificially by a red line.

The efficient nonlinearly induced gain was used to initiate a unique laser oscillation process. A 150 fs pulse with an energy of 3 pJ was coupled to the QDash amplifier which was driven at 210 mA (below oscillating threshold) as illustrated in Fig. 2(a). With the pulse off, the amplifier emitted the broad spectrum shown in Fig. 2(c). With the pulse injection, the device reached oscillation threshold and the output spectrum narrowed to a clear single line at 1570 nm (the same wavelength as for the electrically driven oscillations) as seen in Fig. 2(d).

Figure 2(d) is actually an average spectrum measured with an optical spectrum analyzer. The real intensity of the oscillating line is larger by a factor of 10-20 than shown, determined by the duty cycle of the laser oscillations.

These oscillations are obtained for various optical and electrical excitations but their wavelength is fixed at 1570 nm. The pump pulse at 1550 nm initiates a two-photon induced increase in gain at all wavelengths [6, 16]. This indirect process described schematically in Fig. 2(b) provides the excess gain needed to reach laser oscillation at 1570 nm. The long-lived two-photon induced gain, observed in the pump-probe experiments, lasts long enough to enable a few round-trips of the oscillations (about 30 ps in the 1.5 mm long cavity). Thus it is made possible for these steady-state oscillations to stabilize.

The electrical threshold dependence on pump wavelength at a constant pump energy of 3 pJ is described in Fig. 2(e). For pump wavelengths shorter than 1570 nm, the threshold is basically constant at 210 mA. However, for long pump wavelengths, the threshold drops significantly down to 180 mA at 1608 nm. The wavelength dependence of the threshold is consistent with the concept of two-photon excited carrier generation into the hosting barrier with the longer wavelength pulses exciting carriers at energies closer to the oscillating energy

#161161 - $15.00 USD Received 10 Jan 2012; revised 9 Feb 2012; accepted 9 Feb 2012; published 27 Feb 2012

so that relaxation by carrier-carrier scattering is faster [18] and the gain increase is more efficient. Moreover, pumping at long wavelengths causes no carrier depletion in dashes whose ground state energies are higher than the pumping energy (in contrast, pumping at shorter wavelengths depletes the high energy tails of the density of states function so that all QDashes having a lower energy ground states experience a uniform gain reduction [19]).

To clarify the effect further, we measured the spectrally-resolved time response of the oscillating line using the setup shown in Fig. 3(a). The stimulating pulse at 1530 nm was attenuated, after propagation through the device, using a long-pass filter at 1570 nm. The laser line was then detected by a receiver having a bandwidth of 13 GHz and a 40 GHz sampling scope. Figure 3(b) shows the spectral constellation of the pump (embedded artificially at 1530 nm) and the oscillating line which is 40 nm longer. The measured result is shown in Fig. 3(c) where an initial (unresolved) instantaneous gain increase (circled in red) is followed by conventional relaxation oscillations of the laser as it turns on. The relaxation oscillations have a period of ~250 ps corresponding to the stimulated lifetime of the QWell; it is consistent with the time constant measured in the pump-probe the experiment and agrees well with our hypothesis.

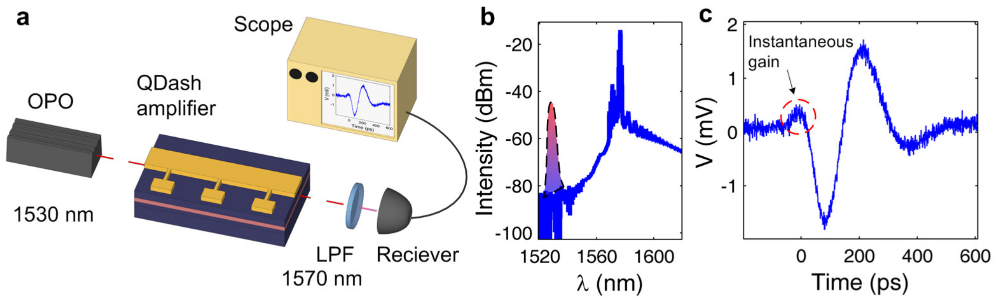

Fig. 3. Spectrally-resolved temporal response. (a) Experimental schematic. (b) Measured oscillating signal with the pump spectral location marked in red artificially. (c) Measured spectrally-resolved temporal response of the oscillating line.

Finally, we examined the signature imprinted on the electromagnetic field of a pulse which induced sufficient gain to reach laser oscillations. To this end, we used a cross frequency-resolved optical gating (X-FROG) system [20] shown schematically in Fig. 4(a). The details of the experimental setup are described in [21]. Utilization of the X-FROG scheme offers two advantages over a standard FROG setup; a higher sensitivity (by more than 30 dB) and the ability to reconstruct an absolute time axis for independently measured fields. Figure 4(b) shows the instantaneous intensity and frequency-shift of the applied Gaussian-like reference pulse at 1585 nm before propagation. X-FROG measurements of two cases were compared. The first is a test case where the amplifier was biased in the absorption regime, at 2.5 mA, and was excited by a relatively weak, 60 pJ pulse. This test case, was used to determine the absolute arrival time of the pulse when no nonlinear processes take place. This low bias measurement was compared with the case of a 190 mA bias and a large pump energy of 925 pJ which initiates laser oscillations. Figure 4(c) shows the X-FROG results for the test case and for the strong pulse. The intensity units in Fig. 4(c) were chosen so that the strong pulse reaches unity at $t = 0$ while the intensity of the test case was artificially enlarged by ~30 times for clarity. The test case pulse maintains its Gaussian-like shape as there are no nonlinear processes that cause pulse distortion. However, the strong pulse transforms into a double-peaked pulse. The arrival time of the first peak coincides with the arrival time of the test case pulse. The second peak is delayed by approximately 250 fs with respect to the first pulse and its intensity is higher. We postulate that the trailing peak originates from the same process that excites the instantaneous gain response [6]. Namely, the leading edge of the pulse induces significant two-photon excited carriers into the high energy levels of the QDash resulting eventually in increased ground state population and hence the gain. The trailing edge

#161161 - $15.00 USD Received 10 Jan 2012; revised 9 Feb 2012; accepted 9 Feb 2012; published 27 Feb 2012

of the pulse senses this gain increase and transforms into the second peak. A delay of about 250 fs is consistent with predictions of carrier–carrier scattering relaxation times [18].

The instantaneous frequency-shift traces also support our hypothesis. During a stimulated emission process, instantaneous frequency of the leading edge reduces, and increases on the trailing edge [22]. The time-resolved instantaneous frequency-shift shown in Fig. 4(c) has a clear inverted double-humped functional form which signifies two independent amplification processes; the first being stimulated emission of the original pulse and the second, a two-photon induced gain response.

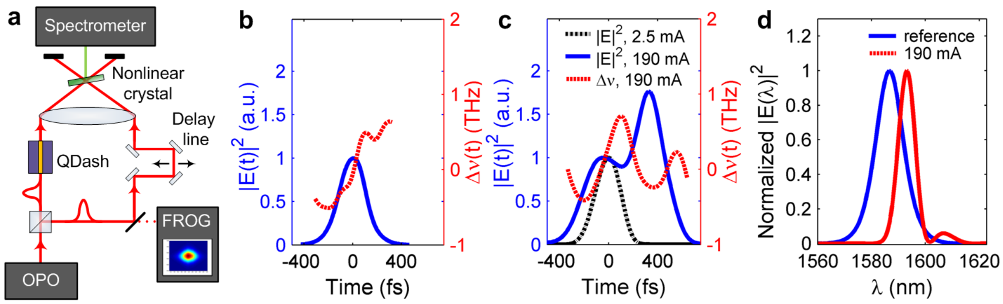

Fig. 4. X-FROG response. (a) Experimental schematic. The pump pulse is filtered using a long-pass filter. (b) Amplitude and instantaneous frequency-shift of the input pulse. (c) Amplitude and instantaneous frequency-shift of the high energy pulse with the amplitude of the test case pulse enlarged artificially for clarity. (d) Input and output average spectra of the high energy pulse.

The instantaneous gain effect is most effective at wavelengths other than that of the stimulating pulse since the carrier depletion is weaker. Examination of the pulse spectral intensity, Fig. 4(d), shows that the pulse red shifts through propagation to unsaturated spectral regions meaning that the instantaneous gain is more efficient thereby further enhancing the self-induced transparency and gain mechanism.

3. Summary

In conclusion, highly efficient nonlinear response was demonstrated in a wire-like QDash gain medium and was employed to initiate unique dynamical responses and laser oscillations. The observations may be due to several processes but we postulate that it originates from highly efficient two-photon excitation. The work sheds new light on the significance and implications of nanostructure-in-a-well type devices, operating in a TPA dominated regime. The characterization of an intense pulse yields a direct measurement of the intra-dash carrier-carrier scattering and testifies to the concept of self-induced amplification. The inherent nonlinearities introduce novel physical concepts and new visions for future applications constituting a major step toward realization of a fully coherent two-photon laser.

Acknowledgments

This work was supported by the Israeli Science Foundation and by the project “GOSPEL” of the European Commission.